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# Temperature decrease and multiple acceleration of structural and phase transformations in metastable metals and alloys under cascade-forming irradiation. Part 1 – General questions and theory

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**Abstract.** Classical radiation physics describes well a number of known phenomena observed under irradiation of metals and alloys (radiation embrittlement, swelling, radiation creep), based on relatively slow processes of thermo- and radiation-enhanced diffusion. Mechanisms based on the description of the defect migration processes can not, however, explain the "low-dose effect" under neutron irradiation and the low-dose "long-range effect" under irradiation with accelerated ions  $E \sim (10^4 - k \times 10^5)$  eV ( $1 \leq k \lesssim 3$ ). The paper is devoted to a brief review of the model that takes into account the nanoscale dynamic effects during cascade-forming irradiation. We are talking about explosive energy release in the regions of dense cascades of atomic displacements (thermal spikes) emitting powerful post-cascaded solitary waves, which can initiate structural-and-phase transformations in metastable media, theoretically, at unlimited distances. The distances at which the effect of accelerated  $(10^4 - k \times 10^5)$  eV ion beams is observed (in the continuous irradiation mode) are sometimes more than a few tens/hundreds of micrometers (at projected ion ranges of less than 1  $\mu\text{m}$ ) and, as recent studies have shown, can reach 1-10 millimeters. These effects are considered on the basis of experimental research data of more than ten different systems. The foundations of the theory of undamped propagation of plane and spherical waves in metastable media are presented. It is noted that the most probable energy of recoil atoms generated by reactor neutrons and fission fragments also belong to the above energy range, which indicates the need to take into account the nanoscale dynamic effects, regardless of the type of the cascade-forming irradiation.

## 1. Introduction

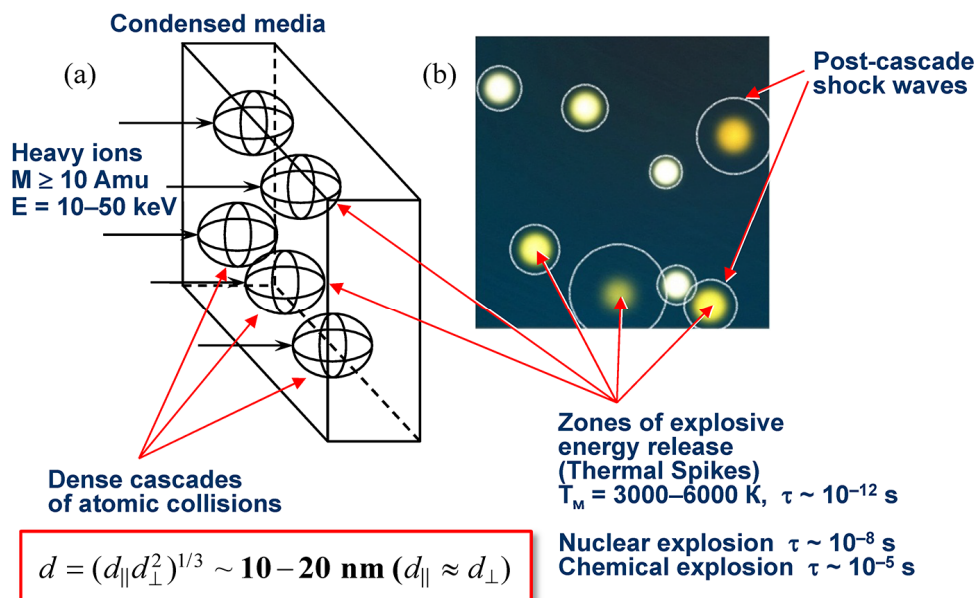
The ion-beam modification of the materials properties has several advantages over traditional processing technologies, namely: (1) an arbitrary chemical element can be introduced into any substance, even if it is insoluble in this substance under equilibrium conditions; (2) the penetration depth of the impurity and its concentration can be strictly controlled; (3) a significantly lower process temperature compared to melting; (4) the ability to form highly defective nonequilibrium surface structures; (5) the environmental cleanliness of the process; and (6) the dimensions of the processed articles remain unchanged.

At the same time, a severe shortcoming of ion implantation is a small ion penetration depth. In many cases this also applies to the thickness of the modified layer, which is insufficient for many practical applications. Nevertheless, it was discovered quite a long time ago that in some cases the change in the properties of materials



is observed at a depth much greater than the range of accelerated ions in matter [1-3]. To explain and predict these effects turned out to be a very difficult task, besides, they can have different physical nature and scale of impact. It has been established that the greatest depth of action is associated with two causes: (1) the formation of high static stresses from the impurities introduced [4-7] (at high fluences), which causes the formation and movement of defects deep into material and (2) nanoscale dynamic effects [8-12], which manifest themselves at extremely low fluences (during just a few seconds of irradiation).

The essence of the latter type of effects lies in the fact that they create (in contrast to, for example, electrons with energy  $>0.5$  MeV) not individual Frenkel pairs, but cascades of atomic displacements, including so called dense cascades, containing up to several tens of thousands of atoms [8, 12] (figure 1). These are areas of explosive energy release. The maximum temperature of the cascade region thermalized during a trillionth of a second can be estimated as the ratio of the energy  $E$  of the accelerated ion (or the primary recoil atom under neutron irradiation) to the number of atoms in the thermalized cascade. Such an estimate is in good agreement with the Monte Carlo calculations [13, 14]. This temperature in the so-called thermal spikes for heavy ions can be from 5000 to 6000 K and higher. The rate of energy release is approximately the same as for a nuclear explosion [12], which leads to the generation of post-cascade shock waves [9, 10, 12].



**Figure 1.** Nanoscale radiation-dynamic effects under cascade-forming irradiation: (a) dense cascades of atomic displacements; (b) the glow of targets (Al, Fe, W etc., see [15, 16]), formation of post-cascade shock waves.

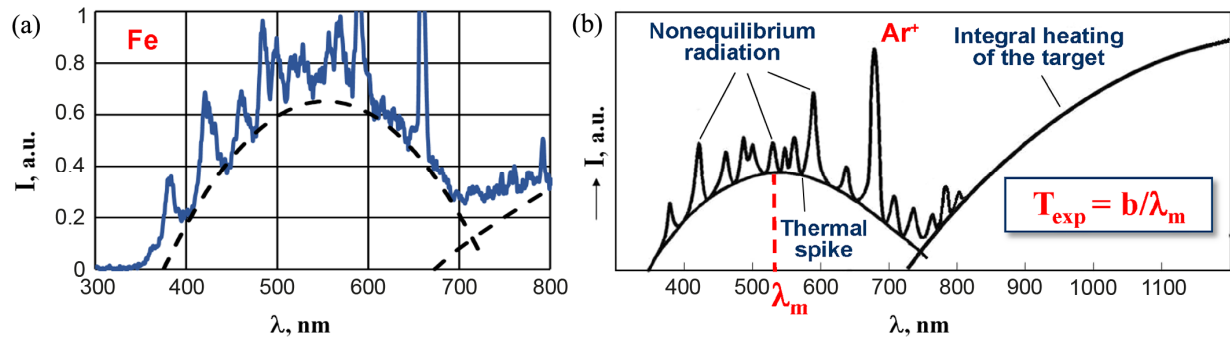
## 2. Experimental evidence of the formation of thermal spikes

For the purpose of experimental evaluation of the temperature of thermal spikes, the velocities of neutral atoms emitted by thermal spikes located close to the target surface are measured. For this, time-of-flight techniques are used, which, unfortunately, have low accuracy for a number of reasons. Nevertheless, the data [17] obtained for the iron target irradiated with  $\text{Ar}^+$  10 keV ions turned out to be close to the calculated [13, 14]. The corresponding temperature in the case of implantation of  $\text{Ar}^+$  ions with an energy of 10 keV into pure iron was 5800 K.

Recently, temperatures of thermal spikes in metal targets (Fe, Zr, W, etc.) have been measured on the basis of a spectral analysis of the glow of these targets during their bombardment with  $\text{Ar}^+$  ions with an energy of 5-20 keV [15, 16] (figure 2). It was found that the continuous emission band in the visible region is due to the emission of thermal spikes, whereas the broad band in the IR region (see the scheme, figure 2b) indicates the integral heating of the target due to the distribution of the energy of thermal spikes in its volume. The measurements mainly reflect the state of the most heated thermal spikes at the initial stage of their life, which gives the maximum contribution ( $E_T$ ) to the spectral radiation density:  $E_T \sim T^4$ .

None of the theoretical models proposed so far (all of them refer to different types of luminescence) can explain all the features of the emission spectrum, in particular, describe the continuous component of the

spectrum (figures 2ab) [18]. In [15, 16] we associate the presence of a continuous band of glow with the formation of thermal spikes, whose temperature is in good agreement with the theoretical calculations for the case of bombardment of Al and Fe by  $\text{Ar}^+$  ions.



**Figure 2.** Spectral composition of Fe target glow under  $\text{Ar}^+$  15 keV irradiation [16]: (a) experiment ( $T=5300$  K,  $T_{\text{target}}=530$  K); (b) a diagram illustrating the formation of the glow spectral composition.

In table 1, the temperatures  $T$  (measured in [15, 16]) and the thermal pressures  $P = 3/2 \Gamma k n T$  [19] in the region of the thermal spikes for pure Al and Fe upon irradiation of these metals by 10 keV  $\text{Ar}^+$  ions are given as an example. Here,  $\Gamma$  is the Gruneisen coefficient ( $\Gamma \approx 2$  for many metals in the condensed state and  $\Gamma = 3/2$  for ultrahigh temperatures, for a monatomic gas),  $k$  is the Boltzmann constant, and  $n$  is the atomic density of the target. It can be seen that the pressure in the region of thermal spikes can exceed the theoretical yield strength of the metal, as in the case for aluminum. Giant pressures arising in the regions of dense cascades of atomic displacements cause the emission of intense elastic or shock solitary waves by thermalized cascade regions [9, 10, 12]. These waves can radically rearrange metastable media, initiating self-propagating structural-phase transformations in the volume of matter [11, 12]. At the front of such waves, the target material can flow as a liquid [12, 17].

### 3. Justification of the possibility of initiating self-propagating structural-and-phase transformations in metastable media under cascade-forming types of irradiation

Undamped propagation of post-cascade waves in metastable media was considered in [11, 12] using the hydrodynamic model [12] (numerical simulation), as well as the phenomenological approach [11, 12]. In the latter case, it is possible to obtain analytical criteria for the possibility of an undamped wave propagation regime (with the transformation of the medium on its front), depending on the control parameters. Unfortunately, the solution for a spherical wave is not given in [12], which raises some doubts about the possibility of such a regime. In this connection, we give a brief derivation and comparison of the relations for the amplitudes of plane and spherical solitary waves in a metastable medium. Such a medium does not exist in the global (absolute), but in some local (intermediate) energy minimum. This is a less deep minimum and it is separated from the global minimum by the energy barrier  $\Delta f$ .

**Table 1.** Temperatures and pressures in thermal spikes, estimated from the emission spectra of Al and Fe in the process their irradiation with  $\text{Ar}^+$  ions,  $E=10$  keV.

$T, K \text{ \& } P, \text{ GPa}$		Experimental yield strength, GPa [16]	Theoretical yield strength, GPa [16]
$T_{\text{exp}} = b/\lambda_m$	$P$	$\sigma_T^{\text{real}}$	$\sigma_T^{\text{theor}} = \left( \frac{\mu}{6} - \frac{\mu}{30} \right)$
Al			
4700	4.89 (3.67)*	0.05-0.120	3.46
Fe			
5800	8.74 (6.56)*	0.1-0.25	9.33

\*/ The pressure in the high-temperature limit ( $T \rightarrow \infty$ ) is indicated in parentheses.

The potential barrier  $\Delta f$  can be overcome by the energy fluctuation, after which such a process in a metastable medium can proceed spontaneously with the release of energy. The necessary energy can also be transferred to some critical volume of matter from the outside, for example, as a result of the formation of thermal spikes during cascade-forming irradiation. Such irradiation, leading to the emission of the above-mentioned powerful elastic or shock solitary waves [9-12], plays the role of a trigger mechanism.

As a result of overcoming the potential barrier, the energy  $\Delta F' = -\Delta F = F_1 - F_2 > 0$ , exceeding  $\Delta f$ , is released.

If the rate of dissipation of wave energy during its propagation in a metastable medium is compensated by the rate of energy release as a result of structural-and-phase transformation at its front, such a wave becomes self-sustaining (constant) in amplitude.

In [12], the propagation in a metastable medium of a solitary compression wave of a rigid profile was analyzed. The usual damping equation:  $d\varepsilon/d\xi = -2\beta\varepsilon$  was replaced by the equation

$$d\varepsilon/d\xi = -2\beta\varepsilon + \Delta F'/(kG), \quad (1)$$

which takes into account the energy release at the front of the wave, which initiates the transformation of the metastable medium into a state with a smaller free energy, where  $\varepsilon$  is the energy at the maximum of the solitary wave profile (per atom or molecule of the medium),  $\xi = x$ ,  $\beta = \delta v$  for a plane and  $\xi = \rho$ ,  $\beta = \delta v + 1/\rho$  for a spherical wave ( $x$  and  $\rho$  are the wave front coordinates,  $\delta$  is the energy absorption coefficient,  $v$  is the wave velocity),  $k$  and  $G$  are the shape coefficient and width of the compression wave profile at half height in the case of a Gaussian wave profile  $k = [\pi/4 \ln(2)]^{1/2} \approx 1.06$  [12]).

Equation (1) is a differential equation of the type

$$y' + P(\xi)y = Q(\xi) \quad \left( y' = \frac{dy}{d\xi} \right). \quad (2)$$

The general solution of such an equation has the form [20]:

$$y = e^{\int P(\xi)d\xi} \left[ \int Q(x)e^{\int P(\xi)d\xi} d\xi + C \right], \quad (3)$$

$y = \varepsilon$ ,  $y' = \varepsilon'$ ,  $P(\xi) = 2\beta$ ,  $Q(\xi) = \Delta F/kG$ ,  $\xi = x$  in the case of a plane wave and  $\xi = \rho$  in the case of a spherical wave.

For the case of a plane wave, the following solution was obtained in [12]:

$$\varepsilon(x) = \begin{cases} \varepsilon_0 \exp[-2\delta(x - x_0)/v], & \varepsilon_0 < \Delta f, \\ \varepsilon^* - (\varepsilon^* - \varepsilon_0) \exp[-2\delta(x - x_0)/v], & \varepsilon_0 \geq \Delta f, \end{cases} \quad (4)$$

where  $\varepsilon^* = \Delta F'/(2\delta kG)$ .

Integration from  $\rho_0$  to  $\rho$  (for  $\rho_0 = \rho$ ,  $y = C = \varepsilon_0$ ) for the case of a spherical wave at  $\varepsilon_0 \geq \Delta f$  yields the following result:

$$\varepsilon(\rho) = \frac{v\Delta F}{2\delta kG} + \left( \varepsilon_0 - \frac{v\Delta F}{2\delta kG} \right) \left( \frac{\rho_0}{\rho} \right)^2 e^{\frac{2\delta}{v}(\rho - \rho_0)} - \frac{v\Delta F}{2\delta kG} \left\{ \frac{1}{\rho^2} \frac{v}{\delta} \left[ \left( \rho - \frac{v}{2\delta} \right) - \left( \rho_0 - \frac{v}{2\delta} \right) e^{\frac{2\delta}{v}(\rho - \rho_0)} \right] \right\} \quad (5)$$

Solutions (4) and (5) indicate that for  $\varepsilon_0 < \Delta f$ , the usual wave attenuation takes place. For  $\varepsilon_0 > \Delta f$ , and simultaneously satisfying the condition  $\varepsilon^* = \Delta F'/(2\delta kG) > \Delta f$ , an autoregulated amplitude wave is formed (restoring its amplitude in the case of its perturbations on inhomogeneities of medium). Analysis of the solutions for other relations of the control parameters  $\varepsilon_0$ ,  $\Delta f$  and  $\varepsilon^*$  is also not difficult. Using the estimate obtained above for the rate of damping of a spherical after-cascade wave in a stable medium (corresponding to a distance of about 100 nm), and also taking into account that the width of the profile of a solitary wave at half-height is, according to [12], of the order of 1 nm, we can obtain that the condition  $\varepsilon^* > \Delta f$  is already satisfied, approximately, for  $\Delta F' > 0.02\Delta f$ .

Substituting the coordinates of the wave front ( $x = vt$  and  $\rho = vt$ ) into equations (4) and (5), we obtain the dependence of the "amplitude" of the compression wave (energy per atom of the medium in its maximum) as a function of time.

The obtained ratios indicate that each solitary wave can initiate the transformation in a metastable medium that propagates theoretically for infinitely large distances. However, the attenuation of the amplitude to the values  $\varepsilon_0 < \Delta f$  can occur on medium defects. So in the paper [21] by the using molecular dynamics method it is

shown that a solitary compression shock wave, when crossing the grain boundary, loses up to 20% of its energy, which can cause a limited spread of processes initiated by solitary waves in metastable media.

#### 4. Conclusion (and the announcement of Part 2 of the review)

This Part 1 of the review presents experimental evidence for the formation of explosive energy release zones – the so-called "thermal spikes" for cascade-forming types of irradiation, and a simple model that predicts the possibility of a cardinal reconstruction of metastable media by powerful post-cascade waves is presented.

In the next Part 2 of the review, as a justification for the validity of the theoretical conclusions, data will be presented on the multiply accelerated transformations in metastable metals and alloys that occur at anomalously low temperatures during ion bombardment, as compared with thermally activated processes. The depth of the transformed zone in metal targets is measured by macroscopic scales, reaching several millimeters, with nanoscale ranges of ions (of low and medium energies) in matter.

Part 2 also gives examples of the modification of the properties of various grades of industrial alloys, as well as the results of direct observation of thermal spikes in the form of traces of reflow on the surface of metallic nanowires.

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